Driven by a guiding principle, the $\lambda^2$ energy scaling law of strong-field phenomena [1], where $\lambda$ is the wavelength of the driving pulses, high-power femtosecond lasers in 3–10 $\mu$m [mid-IR (MIR)] region [2–26] attracts much attention as next-generation sources for ultrafast science and its related areas. One of the most successful lasers of this kind is the relatively low-power but high-pulse-energy KTA optical parametric amplifier (OPA) in the 4-$\mu$m band, which is the longest wavelength band accessible with oxide-crystal-based OPAs. In fact, the 3.9 $\mu$m KTA OPA with 0.16 W and 8 mJ [2] was developed to realize 1.6-keV high-harmonic generation (HHG) [1], laser-plasma x-ray generation [27], and MIR filamentation [28]. Another consequence of this scaling law, whose principle is in the criteria of nonperturbative phenomena, Keldysh parameter $\gamma=\sqrt{T_p/2U_p}\propto\lambda^{-1}J^{-1/2}$ ($I_p$: ionization potential, $U_p$: ponderomotive potential, $J$: intensity of the laser pulses), is the realization of novel nonperturbative phenomena in fragile and functional materials such as semiconductors [3,7,29–31], biomolecules [32], etc.; with MIR pulses, one can realize $\gamma\lesssim1$ while keeping the laser intensity $I$ below their damage threshold.

While the latter applications started to offer novel methodologies for material science and biophysics, the required laser performance is unique and extremely high; both high-pulse energy (210 $\mu$J, to realize the strong-field regime) and $>W$-class high-power (huge number of trials in strong field to detect rare events [24] or small yet statistically significant changes in electric fields [32], etc.) are crucial. However, thermal effects by or coupled with one-/multiphoton processes (e.g., green-induced infrared absorption in MgO:LiNbO$_3$ [33], photorefractive effect in LiNbO$_3$, LiTaO$_3$ [34], and MgO:PPLN [10], and pyroelectrically induced photorefractive damage in MgO:LiNbO$_3$ [35]) in the typical oxide-type nonlinear crystals become serious in developing these lasers [10], while the multiphoton effects are significantly reduced for nanosecond OPAs/optical parametric oscillators (OPOs) [36,37]. Recently, some lasers of this class started to be reported in the 3-$\mu$m-band with niobate-based [8,9,11] or KTA-based [13,22,23] OPAs. For this $>W$-class and sub-100-fs regime, however, elongating their wavelength to its limit, the 4-$\mu$m band, has been a subject of the next challenge, while Ref. [15] reports 105 fs, 0.55 W OPA at 4 $\mu$m and Ref. [20] reports 7.7-W output power before compression with Fourier transform limit (FTL) duration of 140 fs at 3.8 $\mu$m.

We demonstrate a carrier-envelope phase (CEP)-stable, KTA (KTiOAsO$_4$)-based optical parametric amplifier (OPA) delivering 6-cycle (79 fs), 3.8-$\mu$m pulses at a 100-kHz repetition rate with an average power of 4.5 W. The pivotal achievement is the stable generation of supercontinuum (SC) seed pulses in YAG with 1.4-ps pulses, which are the longest for SC generation on its longer wavelength side to 2.2 $\mu$m. This technology offers a robust and simplified OPA architecture with characteristics of passively stabilized CEPS, simplified dispersion management with bulk materials, wavelength tunability for 1.3–4.5 $\mu$m for spectroscopy, and a proof of concept on its power scaling with kilowatt-class pump lasers. The total output power of 17 W (signal plus idler) is achieved, and this high photon flux opens novel statistics-driven spectroscopy for rare events in strong-field physics. © 2019 Optical Society of America

https://doi.org/10.1364/JOSAB.36.002407
Another challenge of MIR OPA development is to clarify the required pump duration for the supercontinuum (SC) seeding scheme [3,5,7,11,17,38–41], which simplifies OPA architecture dramatically. While the typical pulse durations of the pump lasers for MIR OPAs such as Yb-, Nd-, and Ho-based lasers are rather long (≥1 ps) enough for SC generation (SCG) to be interfered with by the competing avalanche ionization in bulk [42,43], this scheme can offer a simplified system without broadband oscillators or pump-seed-synchronization. Similar to the history of Ti:Sa-driven OPAs [44], enormous efforts have been devoted to have/utilize stable SC with these long duration pulses. The present cutting-edge results are (i) a visible pump-seed-synchronization. Similar ing scheme [3,5,7,11,17,38–41], the required pump duration for the supercontinuum (SC) seed

2. OPTICAL LAYOUT AND EXPERIMENTAL RESULTS

Figures 1(a)–1(d) show the optical layout composed of a Yb:YAG thin-disk amplifier (modified TruMicro 5070, Trumpf Scientific Lasers GmbH + Co. KG) weakly seeded by (a) a Ti:Sa oscillator with a Yb:fiber amplifier, (b) a 4D-beam locking system (Alignia, TEM Messtechnik GmbH), (c) a three-stage KTA-based collinear OPA, and (d) an f-2f interferometer with a CEP modulator based on a stepper motor-driven delay stage. Here KTA crystals were adopted as the nonlinear OPA crystals from their proven performance on high-pulse-energy operation and broadband phase-matching in the 4-μm band [2,15,27,28], while their performance in the W-class and sub-100-fs regime is not trivial (e., for the effect of Raman-active phonon modes in KTA, see Ref. [53]). The thin-disk laser delivers 1.3 mJ (130 W) at 100 kHz on target, and its pulse duration was measured with a BBO (BaB₂O₃)-based second-harmonic generation (SHG) autocorrelator to be 1.4 ps [Fig. 2(a)]. After reducing their beam-pointing fluctuation [Fig. 1(b)], mainly from the thermal drift and air turbulence during beam delivery for 4 meters, a portion of the output pulses with 20 μJ was used to generate stable
of the stages were set to profile [Figs. 1(e) and 4], the pumping peak intensities for all waves after OPA1 and OPA2 with dichroic mirrors and filters to avoid undesired interference in their next OPA stages. And to avoid the thermal distortion of the beam observe any amplified superfluorescence when the seed pulses to avoid undesired interference in their next OPA stages.

SC up to \( \approx 2.2 \, \mu \text{m} \) in a 10-mm-thick YAG crystal [Fig. 2(b)]. Here we carefully optimized SCG with a relatively loose focusing geometry and a hard-edge aperture (numerical aperture \( \approx 0.025 \)) and confirmed its stability [Fig. 2(b), 2.2% at 1.41 \( \mu \text{m} \), comparable to that of the pump laser] with longer-duration pulses up to \( \approx 1.6 \) ps. With this careful optimization, the YAG crystal for SCG has not been damaged for 10 months.

The full temporal coherence of the SC was confirmed by its amplification with this OPA with ultrabroad wavelength tunability presented in Fig. 3(c); the typical spectra used in the amplification with this OPA with ultrabroad wavelength tunability [Fig. 2(b), 2.2% at 1.41 \( \mu \text{m} \)]. Here the pumping energy for both stages was 120 \( \mu \text{J} \), and the output pulse energy of the 1.41-\( \mu \text{m} \) signal from OPA2 was 5.0 \( \mu \text{J} \). In OPA3 with a 5-mm-thick Type II KTA with the same phase-matching angles, these 1.41-\( \mu \text{m} \) pulses were injected as seeds, resulting in the generation of 125-\( \mu \text{J} \), 1.41-\( \mu \text{m} \) signal pulses and 45-\( \mu \text{J} \) (4.5-W), 3.8-\( \mu \text{m} \) idler pulses with a 2.0% fluctuation [Figs. 5(a) and 5(b)], which was mainly from that [2.0% SD (standard deviation)] of the pump pulses. This means that OPA3 was successfully in the saturation regime, and this aspect was also confirmed by the fact that the total output power was the same when a 10-mm-length KTA was applied in the high-gain stage OPA1. The pumping power for OPA3 was set to be 90 \( \text{W} \); hence, the total MIR conversion efficiency was 13% for the total output power of 17 \( \text{W} \), which is similar to those of other KTA OPAs [14% for 0.7 \( \text{W} \) [2] and 12.5% for 0.75 \( \text{W} \) [5]]. Here we used relatively thick crystals to compensate the relatively small effective second-order nonlinear coefficient of KTAs \( d_{eff} = 2.01 \, \text{pm/V} \), but we did not observe any amplified superfluorescence when the seed pulses were blocked. And to avoid the thermal distortion of the beam profile [Figs. 1(e) and 4], the pumping peak intensities for all of the stages were set to 57 \( \text{GW/cm}^2 \), which is a value similar to that reported in Ref. [10] and was 20% below the damage threshold of the AR coatings. We also carefully removed idler waves after OPA1 and OPA2 with dichroic mirrors and filters to avoid undesired interference in their next OPA stages.

The CEP of the idler was passively stabilized [52] through DFG at OPA3 between the pump and the 1.41-\( \mu \text{m} \) seed pulses from the same origin, and its spectrum is temporally selected to the slightly shorter wavelength side to avoid the dispersion and absorption by KTA itself and CO₂ in the air around 4255 nm [Fig. 3(b)]. The stability of the CEP of the idler was measured with an inline \( f-2f \) interferometer [Fig. 1(d)]. The 3.8-\( \mu \text{m} \) pulses

![Fig. 2.](image)

(a) Typical autocorrelation trace of the thin-disk laser outputs. Autocorrelation width \( \tau_{AC} \approx 1.95 \) ps, which corresponds to the pulse duration of 1.4 ps under the assumption of Gaussian pulses. The side wings originate from incompressible higher-order dispersion from the CFBG. (b) A typical spectrum of the SC generated in a 10-mm-thick YAG crystal and its excellent stability. Here the integration time for each spectrum is 1 ms, and the error bars indicate the SD of the 1000 spectra. The SDs of the power and the pulse energy at 1.41 \( \mu \text{m} \) were 2.2% (pump 2.0%) and 5.2% (pump 3.7%), respectively. A typical beam profile of the SC taken with a visible camera is also shown in the inset.

![Fig. 3.](image)

(a) Typical spectra of the signals from OPA1 (0.2 \( \mu \text{J} \)), OPA2 (5.0 \( \mu \text{J} \)), and OPA3 (125 \( \mu \text{J} \)). Here the bandwidth of signal waves from OPA1 and OPA2 was limited by phase-matching bandwidth of KTA, and that of idler at OPA3 was manipulated by controlling temporal overlap between the seed and pump in OPA3. (b) A typical idler spectrum at OPA3 (45 \( \mu \text{J} \)), blue curve) measured with acousto-optic-modulator-based MIR spectrometer (MOZZA, Fastlite). The purple and dark yellow curves with broader spectra were obtained at different pumping timings with the same phase matching. (c) Wavelength tunability of the laser system to prove the perfect temporal coherence of the SC pulses and for ultrafast spectroscopy, which requires relatively narrowband pulses (\( \approx 100 \) fs) for selective excitation with \( > \text{W} \) power. The signal covers from 1.3 to 2.0 \( \mu \text{m} \), and the idler covers from 2.1 to 4.5 \( \mu \text{m} \). Here the horizontal scales of (a)–(c) are reciprocal for easier comparison of their energy bandwidths.
drove SCG in a 3-mm-thick ZnSe crystal, which spanned 2 to 5 μm, and its 4.8-μm component was frequency-doubled in a 1-mm-thick Type I GaSe crystal (θ = 10.8°) and interfered with the shorter component of the SC around 2.4 μm. The amplitude ratio of fundamental to second harmonic (SH) pulses was adjusted by rotating a polarizer and the third harmonic (TH) of the fundamental beam generated in the ZnSe crystal was blocked by a long-pass filter. Figure 5(c) shows typical CEP shifts averaged over 3.7 ms as a function of time, and its SD was measured to be 255 mrad. Here we also confirmed the controllability of the CEP by moving the delay stage between the seed and the pump for OPA3 [Fig. 1(d)] for future applications. While the present OPA architecture is carefully designed to reduce phase noises, by applying an active feedback loop with this delay stage, further CEP stabilization for longer time scales will be realized in the near future.

The last advantage of the SC seeding scheme is the availability to use bulk stretcher(s) and/or compressor(s) for its dispersion management, which minimizes power losses and CEP noises compared to those with, e.g., grating- or prism-based dispersion control. Here we have adopted ZnSe-Brewster plates [20 mm, group delay dispersion (GDD): +8 × 10^3 fs^2] for stretching 1.41 μm seed pulses and Si-Brewster plates [3] (40 mm, GDD: +1.6 × 10^4 fs^2) for compressing the amplified 3.8-μm pulses from OPA3 [Fig. 1(c)]. Intrinsic GDD in SCG was measured to be +8 × 10^3 fs^2, and by considering the phase conjugation between the signal and idler waves at OPA3, the GDD of the 3.8-μm pulses is expected to be negligible. Here both the stretcher and the compressor have >90% throughputs, suppressed B-integrals through lateral expansion in the bulk, and small chirps geometrically induced between the plates.

This simple dispersion control scheme was proved by a MIR SHG-FROG (frequency-resolved optical gating) setup using a 30-μm-thick Type I GaSe (θ = 12.3°) SHG crystal and an InGaAs-based near-infrared (NIR) spectrometer (NIRQuest256, Ocean Optics Inc.). Figure 6 summarizes...
the pulse compression results, and the measured pulse duration was 79 fs (≈6 optical cycles: the same duration as that for 1.6 keV HHG [1] and 2 times shorter than the FTL duration of niobate-based OPA at 3.8 μm [20]), whereas the amplified spectrum supports a 70-fs duration. Here the FTL duration for the broadest spectrum [Fig. 3(b), purple curve] was 49 fs, which was elongated to >100 fs due to the above-mentioned dispersion from KTA and CO₂; its further compression is under investigation.

3. APPLICATION TO HHG IN SOLIDS

The excellent performance of the present laser to detect weak signals was demonstrated by a simple HHG experiment with ZnSe polycrystals. The outputs from the OPA were focused with a lens (f = 10 cm) into the exit surface of the ZnSe to minimize the effect of cascaded χ(2) processes with random phase matching observed in Ref. [31]; the spectra of the generated photons were observed with Si and InGaAs spectrometers. Figure 7(a) shows typical harmonic spectra driven by the 3.8-μm and 3.5-μm pulses generated in a 3-mm-thick ZnSe. One can clearly see harmonics up to the 8th order of the 3.8-μm wave below the bandgap [2.67 eV (464 nm)] as well as the novel two peaks around 250 and 400 nm above the bandgap [Fig. 7(b)]. Considering neither their separation nor the position integer multiples of the photon energy of 3.8-μm pulses, they cannot be explained as simple harmonic peaks. Here by using 3.5 μm pulses, we also observed a wavelength-independent peak at 2.61 eV [red curve in Fig. 7(a)] explained by the luminescence from excitonic states created by interband multiphoton (≥ 9) transitions.

To understand the origins of the above peaks, we calculated the imaginary part of the dielectric function ε₂(ω) = Im[ε(ω)] based on the nonlocal pseudopotential model [54,55]. The positions of the two peaks were identified with those of ε₂(ω), and their physical origins are in the so-called Van Hove singularities of the 2D exciton peak and one of the two 3D exciton ones. Here the latter one is related to the 2.61 eV peak by the spin-orbit splitting with an energy of 0.43 eV. The large ε₂(ω) indicates not only large absorption in the perturbative regime but also the existence of a large number of open quantum paths; in general, it can explain the physical origin of these peaks. In fact, the excitonic peaks below the bandgap have been observed in many materials as a photon emission [29], and combining these findings with ultrafast spectroscopic methodology can serve as another route to observe transient band structures of solids.

4. CONCLUSION AND FUTURE PERSPECTIVE

In conclusion, by using an SC seeding scheme with a 1.4-ps Yb:YAG pump laser, we have developed a CEP-stable KTA OPA delivering 79-fs, 3.8-μm, 4.5-W pulses at a 100-kHz repetition rate. Since this duration (1.4 ps) from Yb:YAG thin-disk amplifiers has been realized even in the strong-gain narrowing regime [49–51] and no thermal- or multiphoton-distortion was observed, power scaling of the present OPA architecture with kilowatt-class amplifiers [47,50] is feasible. The obtained total MIR power was 17 W, and such high photon flux allowed us to detect novel and rare events at the Van Hove singularities in ZnSe statistically. As well as a class of such statistics-driven spectroscopy, one of the next challenges is CEP stabilization of signal waves. Since the present thin-disk laser is seeded with a CEP stable Ti:Sa oscillator, the CEP of the signal waves can be stabilized by managing the phase noise during amplifications in the thin-disk RA and the KTA OPA. By superposing these phase-stable signals, idlers, their SHs, etc., the so-called “perfect wave” for HHG [56,57] and other strong-field phenomena can be realized with unprecedented statistics-driven sensitivity for rare events or small changes in electrical fields in field-resolved spectroscopy [32].


Acknowledgment. The authors are grateful to Marcel Schulze for helpful discussions and Yoonman Lee for his technical support.

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Research Article

Vol. 36, No. 9 / September 2019 / Journal of the Optical Society of America B 2413


